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ACTIVATION ENERGY ASYMPTOTICS APPLIED TO BURNING CARBON PARTICL--ETC(U)
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ABSTRACT

The method of activation energy asymptotics is used to describe the entire history of a carbon particle suddenly immersed in a hot oxidizing ambient. Under appropriate conditions which are established by the analysis the history of the particle is shown to consist principally of a heat-up period during which no significant chemical reaction takes place followed by a period involving diffusion-limited mass loss and terminating in complete particle consumption. Between these two extended periods brief ignition and post-ignition periods are described. The final demise of the particle occurs in a complex extinction period during which all the quantities describing the particle behavior undergo large variations.

AMS (MOS) Subject Classifications: 34E05, 80A25

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Work Unit Number 2 (Physical Mathematics)

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SIGNIFICANCE AND EXPLANATION

Libby and Blake treat the combustion of single carbon particles in quiescent ambients with various compositions. Although their analysis is applicable to a variety of conditions, those encountered in entrained flow combustion are emphasized. Accordingly, a cold particle is considered to be injected into a hot ambient, generally involving oxygen but perhaps other active species such as carbon dioxide and water as well. The history of such a particle involves an initial, pre-ignition period during which the particle is heated but undergoes no significant chemical reaction. When the ambient temperature is sufficiently high, the particle reaches a critical, ignition temperature and heterogeneous reactions involving the attack of carbon become effective. This ignition period is followed by post-ignition behavior involving rapid increases in particle temperature which, as a consequence of chemical reaction, generally exceeds that in the ambient. Under the circumstances of practical interest and assumed to prevail in this study both the ignition and post-ignition periods are brief but result in the rate of mass loss by the particle becoming diffusion-limited. There ensues an extended period involving a constant rate of mass loss and terminating in a complex, although brief extinction period during which consumption is complete. The brevity of the transition periods associated with ignition and extinction implies that the principal features of particle behavior we consider are given by a simplified analysis based on inert particle heat-up and diffusion-limited combusion. The present study employs the method of activation energy asymptotics (AEA) to rationalize such an analysis, to establish the conditions for its applicability and to resolve the structure of the transition periods.

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ACTIVATION ENERGY ASYMPTOTICS
APPLIED TO BURNING CARBON PARTICLES

David R. Kassoy* and Paul A. Libby**

INTRODUCTION

Libby and Blake [1-3] treat the combustion of single carbon particles in quiescent ambients with various compositions. Although their analysis is applicable to a variety of conditions, those encountered in entrained flow combustion are emphasized. Accordingly, a cold particle is considered to be injected into a hot ambient, generally involving oxygen but perhaps other active species such as carbon dioxide and water as well. The history of such a particle involves an initial, pre-ignition period during which the particle is heated but undergoes no significant chemical reaction. When the ambient temperature is sufficiently high, the particle reaches a critical, ignition temperature and heterogeneous reactions involving the attack of carbon become effective. This ignition period is followed by post-ignition behavior involving rapid increases in particle temperature which, as a consequence of chemical reaction, generally exceeds that in the ambient. Under the circumstances of practical interest and assumed to prevail in this study both the ignition and post-ignition periods are brief but result in the rate of mass loss by the particle becoming diffusion-limited. There ensues an extended period involving a constant rate of mass loss and terminating in a complex, although brief extinction period during which consumption is complete. The brevity of the transition periods associated with ignition and extinction implies that the principal features of particle behavior we consider are given by a simplified analysis based on inert particle heat-up and diffusion-limited combustion. The present study employs the method of activation energy asymptotics (AEA) to rationalize such an analysis, to establish the conditions for its applicability and to resolve the structure of the transition periods.

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The method of AEA is a recent development in the theory of laminar combustion based on consideration of reaction rates of the form $C\exp(-T_a/T)$ where $C >> 1$ is a nondimensional preexponential factor, T_a is the activation temperature and T is the temperature. Typically $(T_a/T) >> 1$ so that the exponential factor is generally small. To resolve the ambiguity of the value of the product of the two factors, one large and one small, we identify a temperature $T_k = T_a/\ln C$ so that the reaction rate becomes $\exp((T_a/T_k)(1-T_k/T))$; in this form we see the product and thus the reaction rate is small if $T < T_k$, equal to unity if $T = T_k$ and large if $T > T_k$. Thus T_k is exposed as a critical temperature while the reciprocal of the temperature ratio T_a/T_k is an appropriate expansion parameter.

Since its development, AEA has been applied to a variety of problems, e.g., by Law [4], Linan [5], Krishnamurthy *et al.* [6], Peters [7] and Kassoy [8]. A monograph devoted to AEA by Buckmaster and Ludford [9] is near publication. In a recent paper Matalon [10] applies AEA to the description of the departure from chemical equilibrium in the gas surrounding a carbon particle undergoing oxidation. In [2] Libby applies this method to the burning of carbon particles in order to establish a useful ignition criterion and to explain qualitatively the abrupt changes in particle temperature predicted by numerical solutions to the complete equations of thermal response.

BACKGROUND AND ANALYSIS

We start our discussion with the development in [2] based on the more complete analysis in [1]; these references should be consulted for details. It is sufficient for present purposes to describe the basic situation. We consider a spherical carbon particle of initial radius r_{po} suddenly immersed in an oxidizing ambient with temperature T_∞ and with a mass fraction of oxygen denoted $Y_{1\infty}$. Nitrogen treated as an inert diluent is the only other species far from the particle. Only the direct, heterogeneous reaction $C + 1/2\ O_2 \rightarrow CO$ is considered so that the gas mixture involves oxygen, carbon monoxide and nitrogen. More general situations including those involving equilibrium composition in the gas phase, additional heterogeneous reactions and additional species such as hydrogen and water (cf. [3]) can be treated by the methods used here, although increased algebraic complexity is involved. The temperature T_∞ is assumed to be sufficiently high so that ignition takes place.

The Basic Problem and its Asymptotic Treatment

The physical situation described here leads to the following equations which are written in a form appropriate for asymptotic analysis:

$$\theta'(\tau) = \frac{3}{R^2}\left[A\,\theta + D + \phi_r R\,(1-\theta^4)\right] \tag{1}$$

$$R'(\tau) = -\frac{K}{R} \tag{2}$$

$$K = \epsilon \exp\left[-\frac{1}{\epsilon}\left(\frac{\theta_k}{\theta} - 1\right)\right] R\left[(Y_{1\infty} + \tilde{\mu})\, e^{-K} - \tilde{\mu}\right] \tag{3}$$

where

$$A = A(K) = K\left[1 - \tilde{c}_p\,(1-\epsilon^{-K})^{-1}\right]$$

$$D = D(K) = K\left[e^{-K}\,\tilde{c}_p\,(1-e^{-K})^{-1} + E\right]$$

$$E = \tilde{\Delta}_s \left[1 - \frac{\Delta_s}{\tilde{\Delta}_s} \frac{\hat{\mu} - \tilde{\mu}}{\mu'} \right]$$

Equations 1-3 are subject to the initial conditions $\theta(0) = \theta_o$, $R(0) = 1$.

The principal dependent variables in these equations are: $\theta = T_p / T_\infty$, the particle temperature divided by the temperature in the ambient; $R = r_p / r_{po}$, the instantaneous radius divided by its initial value; and $K \geqslant 0$, a nondimensional parameter proportional to the rate of mass loss of the particle. The independent variable τ is a nondimensional time. The other quantities in Eqs. 1-3 are thermochemical parameters, $\tilde{c}_p$, $\tilde{\Delta}_s$, $\tilde{\Delta}$ and the various μ's, are constants of 0(1) related to the thermodynamic and molecular properties of the solid and gas. The quantity $\phi_r > 0$ is a nondimensional parameter related to the radiative properties of the particle and the ambient.

Although Eqs. 1-3 apply to a wide variety of conditions, the numerical examples in the present study involve the following representative values: $\phi_r = 0.186$, $Y_{1\infty} = 0.2$, $\tilde{\mu} = 4/3$, $\tilde{c}_p = 0.583$, $E = 2.18$.

More significant for our considerations is the small parameter ϵ related to the large preexponential factor $\tilde{K}_1$ in the description of the single heterogeneous reaction according to

$$\epsilon\, e^{1/\epsilon} = \tilde{K}_1 \tag{4}$$

where $\tilde{K}_1 >> 1$ involves the product of a constant preexponential factor and of (pr_{po}/η_∞) where p is the pressure and η_∞ is the viscosity of the gas mixture in the ambient. Thus $\tilde{K}_1$ increases directly with increases in pressure and particle size and slowly with decreases in T_∞. Note that conditions of practical interest in entrained flows meet the requirement of $\tilde{K}_1 >> 1$.

The parameter ϵ is also related to a particular value of the nondimensional temperature denoted θ_k which is the critical temperature alluded to earlier and which we shall find is the ignition temperature; the relation is:

$$\epsilon/\alpha = \theta_k = 0(1) \tag{5}$$

where $\alpha \equiv T_\infty / T_a$, the ratio of the ambient and activation temperatures, taken to be fixed.

Several comments regarding Eqs. 4 and 5 are indicated. For a specific calculation of particle behavior the temperatures T_∞ and T_a and thus the parameter α are known. In this connection we note that although the asymptotic analysis is carried out for $\tilde{K}_1 \rightarrow \infty$, $\epsilon \rightarrow 0$, α is a key small parameter determining the applicability of the asymptotic analysis to the specific calculation in question. We note that an alternative asymptotic analysis could involve $\epsilon, \alpha \rightarrow 0$, θ_k fixed implying that both $\tilde{K}_1$ and T_a increase indefinitely. Here we adopt the more applied point of view and imagine that we are interested in the effect on particle behavior of changes in the the factor (pr_{po}/η_∞). In a specific calculation that factor and thus $\tilde{K}_1$ are also known so that Eq. 4 determines ϵ and Eq. 5 determines θ_k which may thus from our adopted point of view be considered a function of $\tilde{K}_1$ and α. Increases in $\tilde{K}_1$ due to increases in the factor (pr_{po}/η_∞) lead to decreases in ϵ and in θ_k. If we associate θ_k with an ignition temperature, we conclude that the temperature at which vigorous heterogeneous reaction is initiated decreases as the overall reaction rate is increased, a physically appealing conclusion. However, it must be noted that Eq. 4 calls for large changes in $\tilde{K}_1$ to achieve relatively small changes in ϵ. To illustrate we show in Table 1 numerical values of ϵ, $\tilde{K}_1$ and θ_k. We observe that a halving of ϵ requires an increase in $\tilde{K}_1$ by nearly four orders of magnitude while Eq. 5 implies an accompanying reduction of θ_k by only one-half with α held fixed.

The relations involving ϵ, $\tilde{K}_1$ and θ_k influence greatly the behavior of the mass loss parameter K via Eq. 3. A consideration of the range of possible values of K is helpful in achieving understanding of the asymptotic analysis and its applicability to a specific calculation. To that end we rewrite Eq. 3 as

$$K = \epsilon \exp\left[\frac{1}{\epsilon}\left[1 - \frac{\theta_k}{\theta}\right]\right] R\,(Y_{1\infty} + \tilde{\mu})\,(e^{-K} - e^{-K_d}) \tag{6}$$

where the special value of K denoted K_d is defined by $\exp(-K_d) = \tilde{\mu}/(Y_{1\infty} + \tilde{\mu})$ and is the diffusion-limited rate of mass loss, the rate determined by the ability of oxygen to diffuse to the surface of the particle independent of the kinetics of the heterogeneous reaction. For the values of the thermochemical parameters used in the numerical examples $K_d = 0.140$.

We now discuss Eq. 6; prior to ignition when $\theta < \theta_k$ (meaning bounded away from θ_k in the limit $\epsilon \to 0$) and $R \approx 1$, Eq. 6 indicates that K is basically an exponentially small quantity and the rate of mass loss is kinetically controlled, i.e., diffusion of oxygen to the surface of the particle provides no effective inhibition to mass loss. Ignition occurs when $\theta_k - \theta = 0(\epsilon)$ so that $K = 0(\epsilon)$. The identification of θ_k with ignition is now clear.

Further increases in θ above θ_k lead to $0(1)$ values of K. For example, when $(\theta_k/\theta) = 1 - -\epsilon \ln(1/\epsilon) + 0(\epsilon)$ and $R = 0(1)$, the exponential term in Eq. 6 is large, of $0(1/\epsilon)$ and $K = 0(1)$. However, the growth of K is limited by the last factor on the right side of Eq. 6, i.e., by $K \to K_d^-$, so that the growth in the magnitude of the exponential term with increasing θ is compensated by decreasing values of the last factor. When $K \to K_d^-$, Eq. 6 can be rewritten in the following illuminating form:

$$K/K_d \approx 1 - \left\{ \epsilon \tilde{\mu} R \exp\left[\frac{1}{\epsilon} \left[1 - \frac{\theta_k}{\theta} \right] \right] \right\}^{-1} \tag{7}$$

Equation 7 applies provided the second term on the right side is suitably small compared to unity. This is the case as $\epsilon \to 0$ provided θ is sufficiently large for the operative order of magnitude of R. For example, if $R = 0(1)$ and

$$\frac{\theta}{\theta_k} = \frac{1}{1 - \epsilon \ln(\beta(\epsilon))} \tag{8}$$

where $\beta >> 0(1/\epsilon)$, e.g., $\beta = \epsilon^{-2}$, $K \approx K_d$. However, as complete consumption of the particle is achieved so that $R = 0(\epsilon)$, then Eq. 7 applies and the rate of mass loss is diffusion-limited only if

$$\frac{\theta}{\theta_k} = \frac{1}{1 - \epsilon \ln(\gamma(\epsilon))}$$

where $\gamma >> 0(1/\epsilon^2)$, e.g., $\gamma = \epsilon^{-3}$. Compared to the result for $R = 0(1)$ we see that the temperature deviation from θ_k must be larger when $R << 1$ if diffusion-limited behavior is to prevail. It should be noted that when $\theta > \theta_k$ and $\epsilon \to 0$, the factor $\epsilon \exp((1 - (\theta_k/\theta))/\epsilon)$ is exponentially large so that R must be exponentially small if $K = 0(1) < K_d$. In contrast we observe from Eq. 7 that for

$R = 0(1)$ $K \to K_d^-$ if $\theta - \theta_k = \epsilon \ln(\gamma(\epsilon)/\epsilon R)$ where $\gamma(\epsilon) >> 0(1)$ and R have an algebraic dependence on ϵ. We thus conclude from Eq. 7 that diffusion-limited behavior occurs for both $R = 0(1)$ and $R = 0(1)$ in an algebraic sense if $\theta - \theta_k = 0(\epsilon \ln \mu(\epsilon))$ provided $\mu(\epsilon)$ is suitably but algebraically large in the limit $\epsilon \to 0$, suitably being determined by the requirement that the second term in Eq. 7 be small compared to unity. Thus only a small deviation from θ_k is required to achieve diffusion-limited mass loss. These arguments represent a generalization of the θ_n- sequence introduced by Libby [2] and used to explain qualitatively the particle behavior predicted by exact numerical solutions to the describing equations.

The question arise as to the applicability of these results related to $\epsilon \to 0$ for a specific calculation involving a finite but small value of ϵ. From a consideration of Eqs. 3 and 7 we cannot be assured that for a particular value of ϵ the actual histories $\theta(\tau)$ and $R(\tau)$ respect the requirements for diffusion-limited behavior predicted by the asymptotic analysis for small increments in θ above θ_k. However, we seek within the asymptotic analysis itself symptoms of failure, i.e., failure to achieve diffusion-limited behavior. An indication of such failure is indicated in Table 2 where we show the dependence of the second term on ϵ and θ for $R = 1$ with the thermochemical values used in the numerical examples.

Note that for our specific value of α, namely $\alpha = 1/9$, the limiting value of ϵ for ignition corresponds to $\theta_k \approx 1$ and is thus equal to 0.111. We see from Table 2 that the range of values of ϵ and θ yielding diffusion-limited behavior in a specific calculation and thus consistency with the asymptotic analysis may call for values of ϵ smaller than are usually found to be acceptable in asymptotic analyses.

The history of a carbon particle suddenly immersed in a hot oxidizing ambient as described earlier can now be reexamined. During pre-ignition $\theta < \theta_k$, $R = 0(1)$ and $K \leqslant \exp(-\epsilon^{-1}(\theta_k/\theta - 1))$, i.e., K is exponentially small. In the neighborhood of $\theta = \theta_k$ abrupt changes take place in θ and K, changes associated with the ignition and post-ignition periods which are found to have durations of $0(\epsilon)$ and $0(\epsilon \ln(1/\epsilon))$. Under appropriate circumstances an extensive period of diffusion-limited behavior can ensue with $\theta - \theta_k > 0(\epsilon \ln(1/\epsilon))$. Although it is characteristic of all of these periods that $\theta'(\tau) > 0$, eventually the right side of Eq. 1 must vanish. Thereafter a complex extinction process evolves in which $\theta'(\tau) < 0$ and $\theta \to 1$.

With the basic situation described we now can take up the details of the analysis. In [2] the pre-ignition and ignition periods are studied by eliminating τ between Eqs. 1 and 2 so that the differential equation $dR/d\theta$ is subjected to asymptotic analysis. In this approach the time variation is found in the form $\tau = \tau(\theta)$ from knowledge of $R = R(\theta)$. Here we find analysis of the particle history prior to extinction is handled more conveniently with τ retained as the independent variable.

Pre-Ignition and Ignition Periods

If $R = 0(1)$ and $\theta(0) < \theta < \theta_k$ so that K and $R-1$ are exponentially small, $R = 1$ and Eq. 1 becomes

$$\theta'(\tau) = 3\,\bar{c}_p\,(1-\theta) + \phi_r\,(1-\theta^4) \tag{9}$$

which describes the thermal response of a spatially homogeneous, inert sphere subject to conductive and radiative heating. A supercritical response, i.e., one involving significant chemical reaction and particle consumption, evolves only if $\theta'(\tau) > 0$. This inequality is met if $\theta_k < 1$ so that we establish one limitation of the asymptotic analysis. For a given value of $\tilde{K}_1$ and thus of ϵ the value of θ_k depends on α; we thus appreciate its important role in determining particle behavior. An inverted quadrature of Eq. 9 gives $\tau = \tau(\theta)$. It follows that the duration of the induction period given by τ_k is known once θ_k is known from the specified values of ϵ and α. The solution of Eq. 9 in the neighborhood of $\tau = \tau_k$ is

$$\theta(\tau < \tau_k) \simeq \theta_k \left[1 + \lambda^2(\tau - \tau_k) + \cdots \right] \tag{10}$$

where $\lambda^2 = 3(\bar{c}_p\,(1-\theta_k) + \phi_r\,(1-\theta_k^4))/\theta_k > 0$ for $\theta_k < 1$ has a known value assumed to be $0(1)$ in the limit $\epsilon \to 0$.

Centered about τ_k is the ignition period during which θ and K increase and R decreases. To describe this period we find that the appropriate scaling transformations are

$$\theta \simeq \theta_k \ (1 + \epsilon \ \hat{\theta}_1 + \ \cdots)$$

$$R \simeq 1 - \epsilon^2 \hat{R}_2 \tag{11}$$

$$\tau \simeq \tau_k + \epsilon \hat{\tau} \ .$$

Then Eq. 3 gives $K = \epsilon \ Y_{1\infty} \exp(\hat{\theta}_1) Y_{1\infty} << 1$ to a first approximation. With $A(K)$ and $D(K)$ appropriately expanded about $\theta = \theta_k$ Eqs. 1 and 2 become

$$\hat{\theta}'_1(\hat{\tau}) \simeq \lambda^2$$

$$\hat{R}'_2(\hat{\tau}) \simeq Y_{1\infty} e^{\hat{\theta}_1} \ .$$

The following solutions which match properly to the pre-ignition solution for θ and to $R = 1$ are readily found to be

$$\hat{\theta}_1(\hat{\tau}) = \lambda^2 \hat{\tau}$$

$$\hat{R}_2(\hat{\tau}) = \frac{Y_{1\infty}}{\lambda^2} \ e^{\lambda^2 \hat{\tau}} \tag{12}$$

The exponential growth of K limits the validity of the expansion in Eq. 11. In particular when $\epsilon \exp(\lambda^2 \hat{\tau}) = 0(1)$, then $K = 0(1)$, $\epsilon^2 \hat{R}_2 = 0(\epsilon)$, $\epsilon \hat{\theta}_1 = \epsilon \ln(1/\epsilon) + 0(\epsilon)$ and $\epsilon \hat{\tau} = \epsilon \ln(1/\epsilon)/\lambda^2 + 0(\epsilon)$. If we define $\hat{\tau}_i = \lambda^{-2} \ln(1/\epsilon)$, then it follows from Eq. 12 that

$$\theta(\tau \rightarrow \tau_i) \simeq \theta_k \left[1 - \epsilon \ln(1/\epsilon) + \lambda^2 (\tau - \tau_i) + \ \cdots \right] \tag{13}$$

$$\tau_i = \tau_k - \hat{\tau}_i$$

Post-Ignition Period

The nonuniformities in the solutions for the ignition period imply that in the next region

$$\theta \simeq \theta_k \left[1 + (\epsilon \ln \frac{1}{\epsilon})\right] + \epsilon\, \tilde{\theta}_1 + \cdots)$$

$$R \simeq 1 - \epsilon\, \tilde{R}_1 + \cdots \tag{14}$$

$$\tau \simeq \tau_i + \frac{\epsilon}{\lambda^2}\, \tilde{\tau} = \tau_k - \frac{\epsilon}{\lambda^2} \left[\ln (\frac{1}{\epsilon}) - \tilde{\tau}\right] .$$

Substitution into Eqs. 1-3 yields

$$\tilde{\theta}'_1 \simeq \frac{3}{\lambda^2 \theta_k} \left[A\, \theta_k + D + \psi_r (1 - \theta_k^4)\right]$$

$$\tilde{R}_1 (\tilde{\tau}) \simeq \frac{K}{\lambda^2} \tag{15}$$

$$K \simeq e^{\tilde{\theta}_1} \left[Y_{1\infty} + \tilde{\mu}) e^{-K} - \tilde{\mu} \right] .$$

It should be noted here that we are considering a particle with a temperature still close to the ignition temperature θ_k and with a radius with $R \simeq 1$. However, the third of Eqs. 15 implies that significant mass loss is encountered because $K = 0(1)$.

As long as the term within brackets on the right side of the first of Eqs. 15 is positive, the temperature variation is monotonically increasing. Accordingly, it worth examining the variation of this factor with K. In Table 3 we give for the thermochemical values used in the numerical examples the variation of the quantities $A(K)$ and $D(K)$. We deduce therefrom that the factor in question is positive over the entire range of K provided that $\vartheta_k < 1$ as required earlier.

Equations 15 must be solved numerically subject to matching to Eq. 13 and to the vanishing of $\tilde{R}_1$ as $\tilde{\tau} \approx -\infty$; thus we require

TABLE 1		
ϵ	$\tilde{K}_1 = \epsilon\, e^{1/\epsilon}$	$\theta_k = \epsilon/\alpha$
0.06	1.04 (10^6)	$0.06/\alpha$
0.08	2.15 (10^4)	$0.08/\alpha$
0.10	2.20 (10^3)	$0.10/\alpha$
0.12	4.99 (10^2)	$0.12/\alpha$

TABLE 2			
		$\left(\tilde{\mu}\,\epsilon \exp\left(\frac{1}{\epsilon}\left(1-\frac{\theta_k}{\theta}\right)\right)\right)^{-1}$	
ϵ	θ_k	$\theta = 1$	$\theta = 1.5$
0.06	0.54	5.9×10^{-3}	2.9×10^{-4}
0.08	0.72	2.8×10^{-1}	1.4×10^{-2}
0.10	0.90	2.5	1.4×10^{-1}

TABLE 3		
K	$A(K)$	$D(K)$
$K \to 0$	-0.583	0.583
0.02	-0.569	0.621
0.05	-0.548	0.678
0.1	-0.513	0.772
$K_d = 0.140$	-0.485	0.848

$$\tilde{\theta}_1\,(\tilde{\tau} \rightarrow -\infty) \simeq \tilde{\tau}$$

$$\tilde{R}_1\,(\tilde{\tau} \rightarrow -\infty) \simeq 0\,.$$

As $\tilde{\tau} \rightarrow \infty$, $\tilde{\theta}_1$ increases in an unbounded fashion and it follows that as a consequence

$$\tilde{\theta}'_1\,(\tilde{\tau} \rightarrow \infty) \sim \omega^2 \equiv \frac{3}{\lambda^2 \theta_k}\left[-A_d\theta_k + D_d + \theta_r\,(1-\theta_k^4)\right]$$

$$A_d \equiv -A\,(K_d) > 0$$

$$D_d \equiv D\,(K_d) > 0$$

so that

$$\theta(\tilde{\tau} \rightarrow \infty) \simeq \theta_k\left[1 + \epsilon \ln\,(\frac{1}{\epsilon}) + \epsilon\,(\omega^2\tilde{\tau} + K_1) + \cdots\right]\,.$$

The present analysis is valid provided $\omega^2 = 0(1) > 0$. In fact for specific thermochemical values we can determine a special value of θ_k denoted θ_k^* which makes ω^2 vanish. Thus we have

$$-A_d\theta_k^* + D_d + \phi_r\,(1-\theta_k^{*4}) = 0$$

For the values used in the numerical examples $\theta_k^* = 1.234$. This value identifies a second limit on the applicability of the present asymptotic analysis to a specific calculation. Physically, θ_k^* is the temperature a particle with $R \simeq 1$ can achieve when it undergoes diffusion-limited chemical reaction. The analysis is self-consistent provided the second term on the right side of Eq. 7 with $\theta = \theta_k^*$ and $R = 1$ is suitably small compared to unity. This requirement defines loosely a maximum value of ϵ; for example, if we permit the second term to be 0.1, ϵ can be as large as 0.085. If we require that term to be 0.01, ϵ can be as large as 0.07. Accordingly, when we compare the prediction of the asymptotic analysis with exact numerical calculations, we take $\epsilon = 0.060$ but to illustrate the nature and extent of the disagreement which results when the asymptotic analysis is overextended, we also present results for

$\epsilon = 0.090$. If ϵ is suitably small, we conclude that once a supercritical event is initiated, particle consumption proceeds with vigor.

The interesting nonuniformity occurs when $\epsilon\tilde{\tau} = 0(1)$. Equations 14 then imply that $\epsilon\hat{R}_1 = 0(1)$ and $\epsilon\tilde{\theta}_1 = 0(1)$. Since $\tilde{\theta}_1$ becomes unbounded, it follows from the third of Eqs. 15 that $K \to K_d$ and that the diffusion limit is achieved while $R = 0(1)$.

Diffusion-Limited Period

The nonuniformity at the end of the preceding period suggests that the appropriate variables for this succeeding period are

$$\theta = \theta(s), \quad R = R(s), \quad \tau = \tau_k + \frac{\epsilon}{\lambda^2} \ln\left(\frac{1}{\epsilon}\right) + \frac{s}{K_d}, \quad \frac{K}{K_d} = 1 - \frac{\exp\left[\frac{-1}{\epsilon}\left(1 - \frac{\theta_k}{\theta}\right)\right]}{\epsilon\, \tilde{\mu}\, R} \tag{16}$$

where $\theta > \theta_k$. It follows that the mass loss is basically constant $K = K_d$ while the dependent variables $\theta(s)$ and $R(s)$ retain their primitive form implying $0(1)$ changes from θ_k and unity respectively. The equation for $R(s)$ can be integrated directly to yield

$$R = (1 - 2s)^{1/2} \tag{17}$$

where we impose the condition $R(s \to 0) = 1$. Equation 1 becomes

$$\frac{d\theta}{ds} = \frac{3}{K_d(1-2s)} \left[-A_d\theta + D_d + \phi_r (1-2s)^{1/2} (1-\theta^4)\right] \tag{18}$$

Equation 18 must be solved numerically subject to the initial condition $\theta(s = 0) = \theta_k$.

We obtain immediately from Eq. 17 an estimate for the extinction time since $R = 0$ when $s = 1/2$; thus

$$\tau_c \simeq \tau_k + (2\,K_d)^{-1} \tag{19}$$

It is interesting to note that the increment added to the induction time depends only on the concentration of oxygen in the ambient and on the molecular weight parameter $\tilde{\mu}$. This implies that particle lifetimes in pure oxygen should be one quarter those in air provided the induction period is relatively brief. The experimental results reproduced in [1] for the lifetimes of a wide variety of carbon containing particles in air and pure oxygen are in accord with this estimate.

Comparison of Exact and Asymptotic Solutions

The combination of the pre-ignition period plus the diffusion-limited period obtained from the solution of Eq. 18 provides the main features of particle behavior according to AEA. At this juncture it is thus appropriate to compare exact numerical calculations with the predictions of the asymptotic analysis. As indicated earlier we do so for two values of ϵ: 0.060 and 0.090. One is suitably small so that the two limits on the applicability of the asymptotic analysis are respected and the second too large for the condition resulting in $K \rightarrow K_d^-$ to apply. However, both values of ϵ with our fixed value of α yield suitable values of θ_k, namely 0.540 and 0.810 respectively. The previously given values of the thermochemical parameters apply.

Figures 1-3 give the variations with τ of the particle temperature parameter θ, of the particle radius in terms of R and of the mass loss parameter K respectively. For the larger value of ϵ, i.e., for the smaller chemical kinetic rate, ignition is delayed, the rate of mass loss is smaller than the diffusion-limited rate and the particle lifetime is increased.

Also shown on these figures are the predictions based on the asymptotic analysis for the two dominant periods of particle lifetime, the pre-ignition and diffusion-limited periods. First we call attention to the response of the inert particle as given by the solution of Eq. 9 and the two values of the ignition time τ_k associated with that response. These predictions are in good agreement with the exact solutions. For the smaller value of ϵ we see that the asymptotic analysis predicts many features of particle

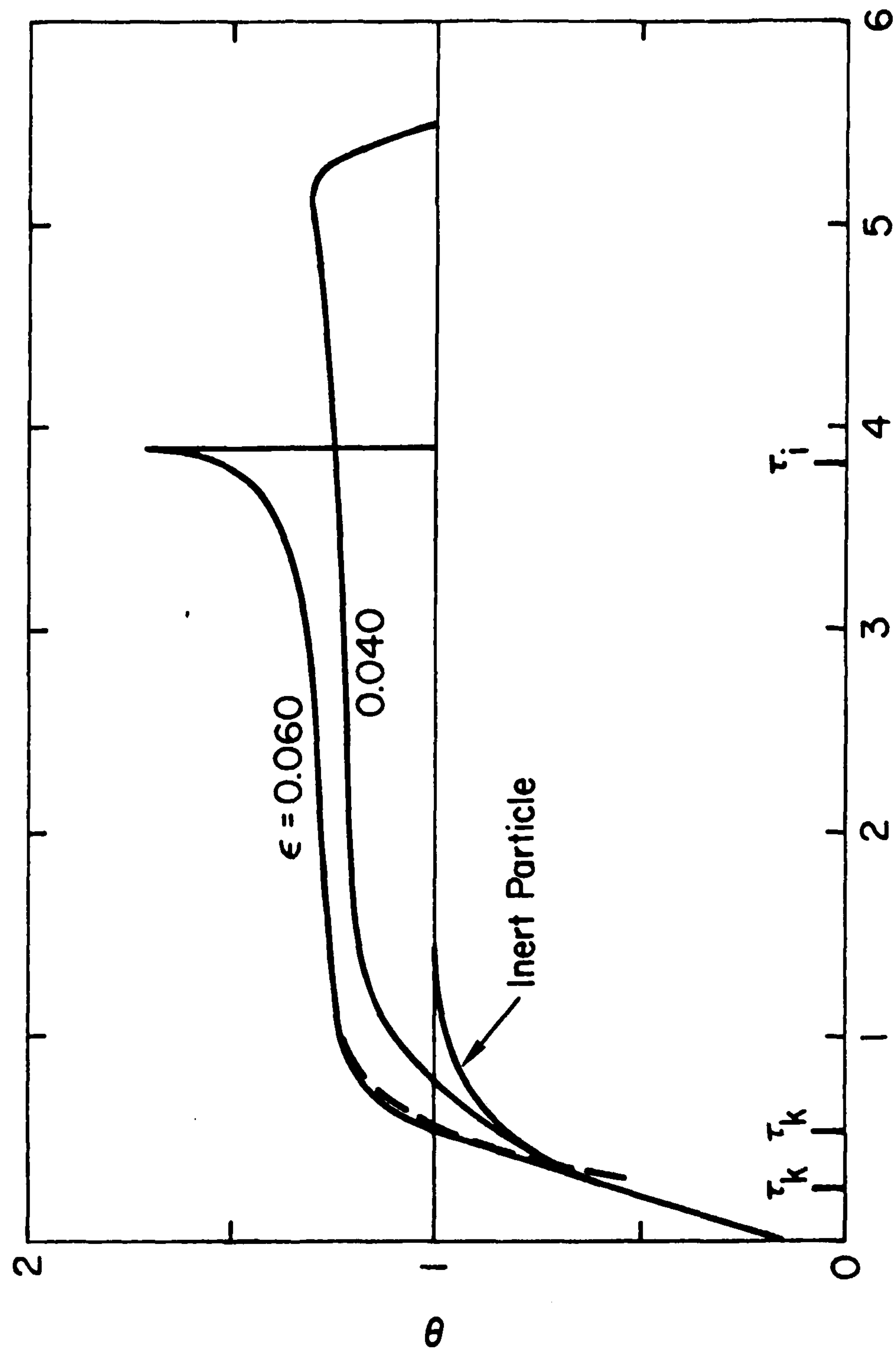

Figure 1. The temperature histories of particles: ———— exact numerical solutions; — — — asymptotic solution for $\epsilon = 0.060$.

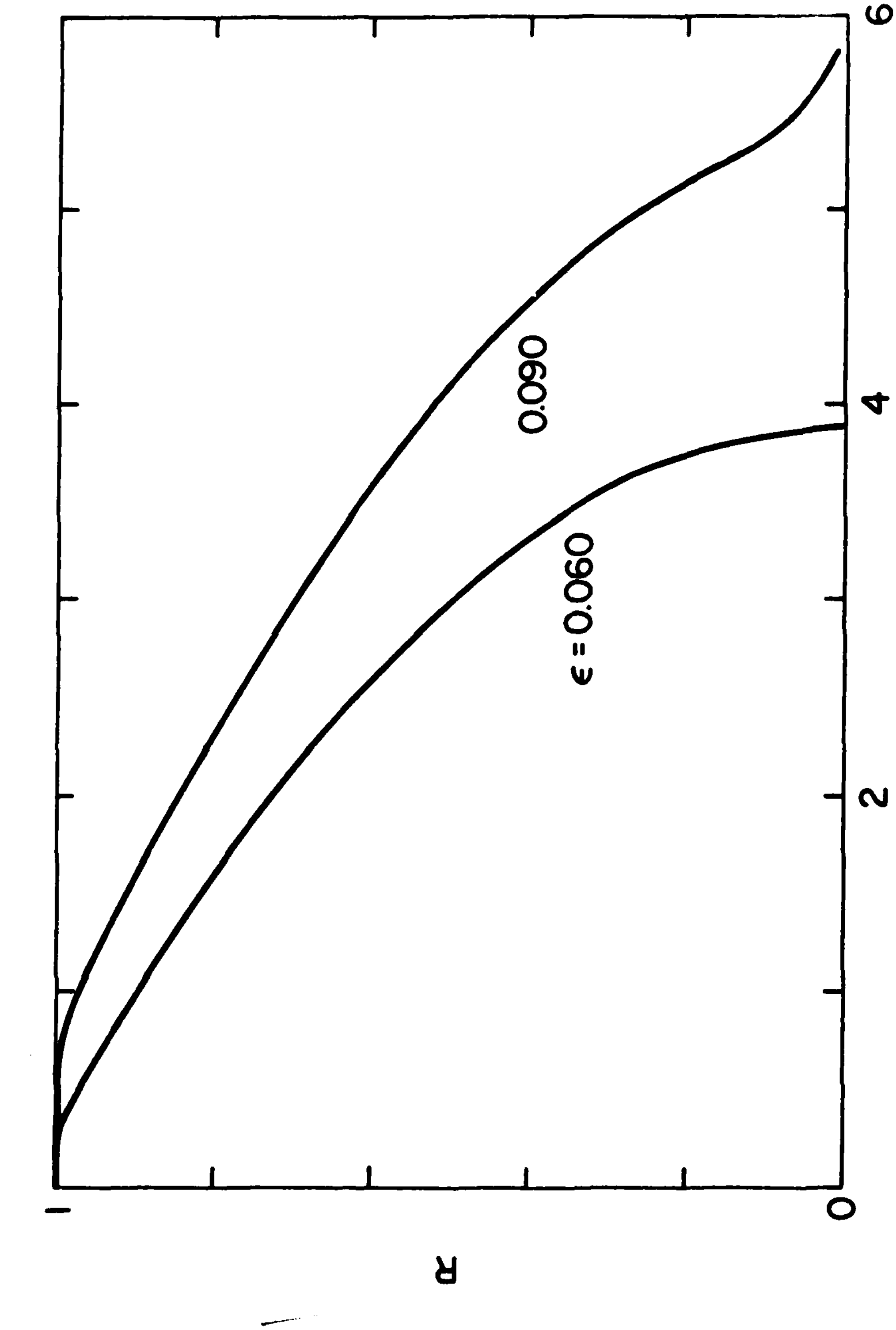

Figure 2. The radius histories of particles as given by exact numerical solutions.

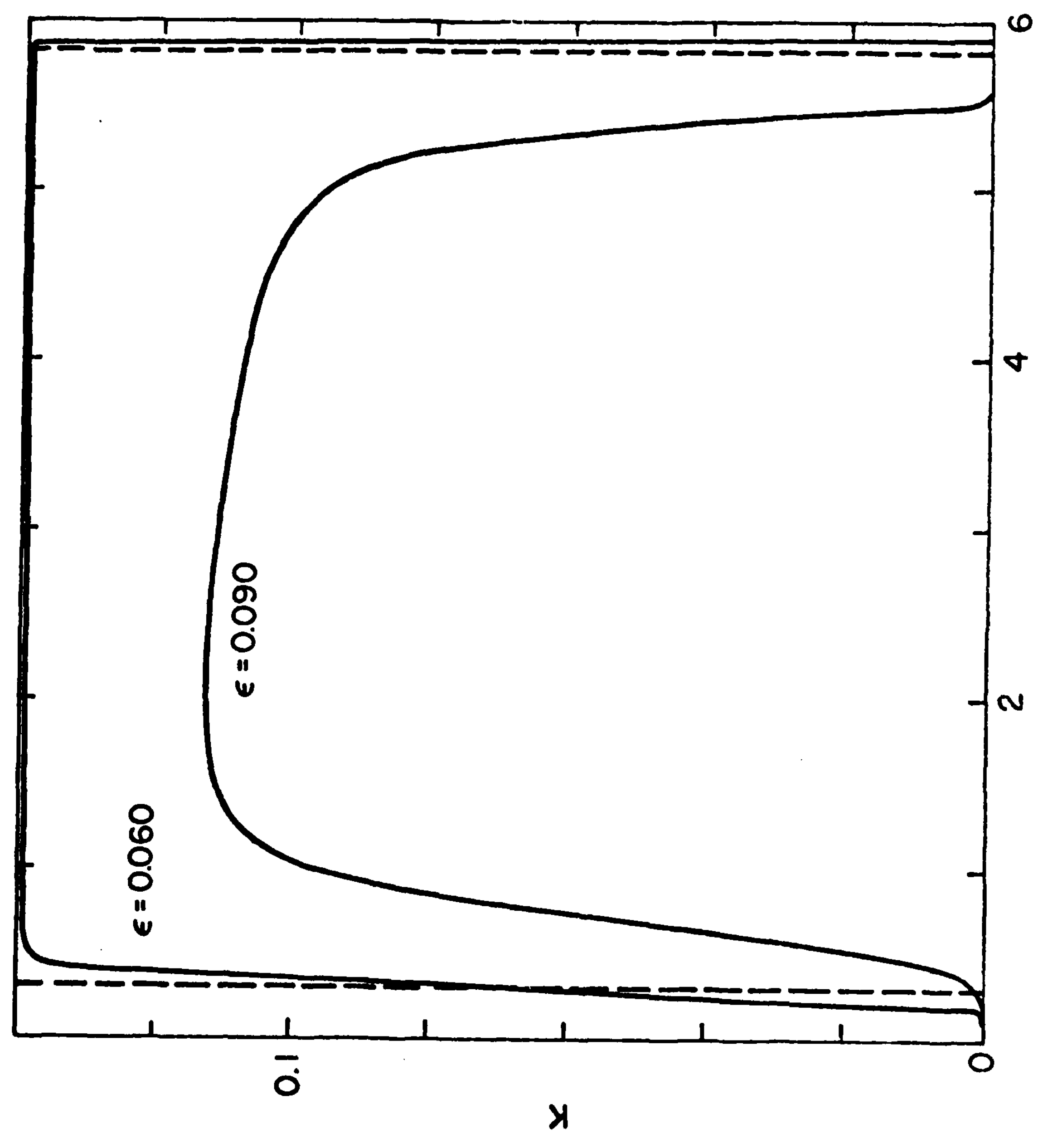

Figure 3. The variation of mass loss parameter (see Fig. 1 for legend).

behavior with an accuracy acceptable for most purposes. This is not the case for $\epsilon = 0.090$. Here the difference between the exact and diffusion-limited rate of mass loss predicted by the asymptotic analysis leads to a considerable error in particle lifetime. It is worth noting that for $\epsilon = 0.060$ the value of K given by the exact solution is slightly less than K_d reflecting both the large but finite magnitude of the exponential factor in Eq. 3 and the small but nonzero value of the second term in Eq. 7.

It will be found convenient in the analysis of the extinction period to obtain solutions in the form $\theta = \theta(K)$ and it is therefore of interest to examine the details of the transition from inert particle behavior ($K \approx 0$) to diffusion-limited behavior ($K \approx K_d^-$) in this form. We do so for both the exact and asymptotic solutions in Fig. 4 for $\epsilon = 0.060$. The satisfactory agreement between the two solutions is noted.

We thus conclude from these results that AEA provides a useful approximate description of particle behavior provided that the reaction rate parameter $\tilde{K}_1$ is sufficiently large for the expansion parameter ϵ to be appropriately small and that the temperature ratio parameter α is similarly small.

The Extinction Period

Although the main results of practical utility are given by the analysis presented to this juncture, the elaborate structure of the extinction period during which $\theta \to 1$ and $R \to 0$ is interesting. It is clear from the form of Eq. 18 that a singularity develops in the θ-solution when $s \to \frac{1}{2}^-$. By elementary means we find that

$$\theta\left(s \to \frac{1}{2}\right) \simeq \theta^* - m\left(\frac{1}{2} - s\right)^{1/2} + \cdots \qquad (20)$$

where

$$m \equiv 3\sqrt{2}\,\phi_r(\theta^{*4} - 1)/(3A_d - K_d) \ .$$

The singularity indicated by Eq. 20 is weak in the sense that θ approaches a finite value while its

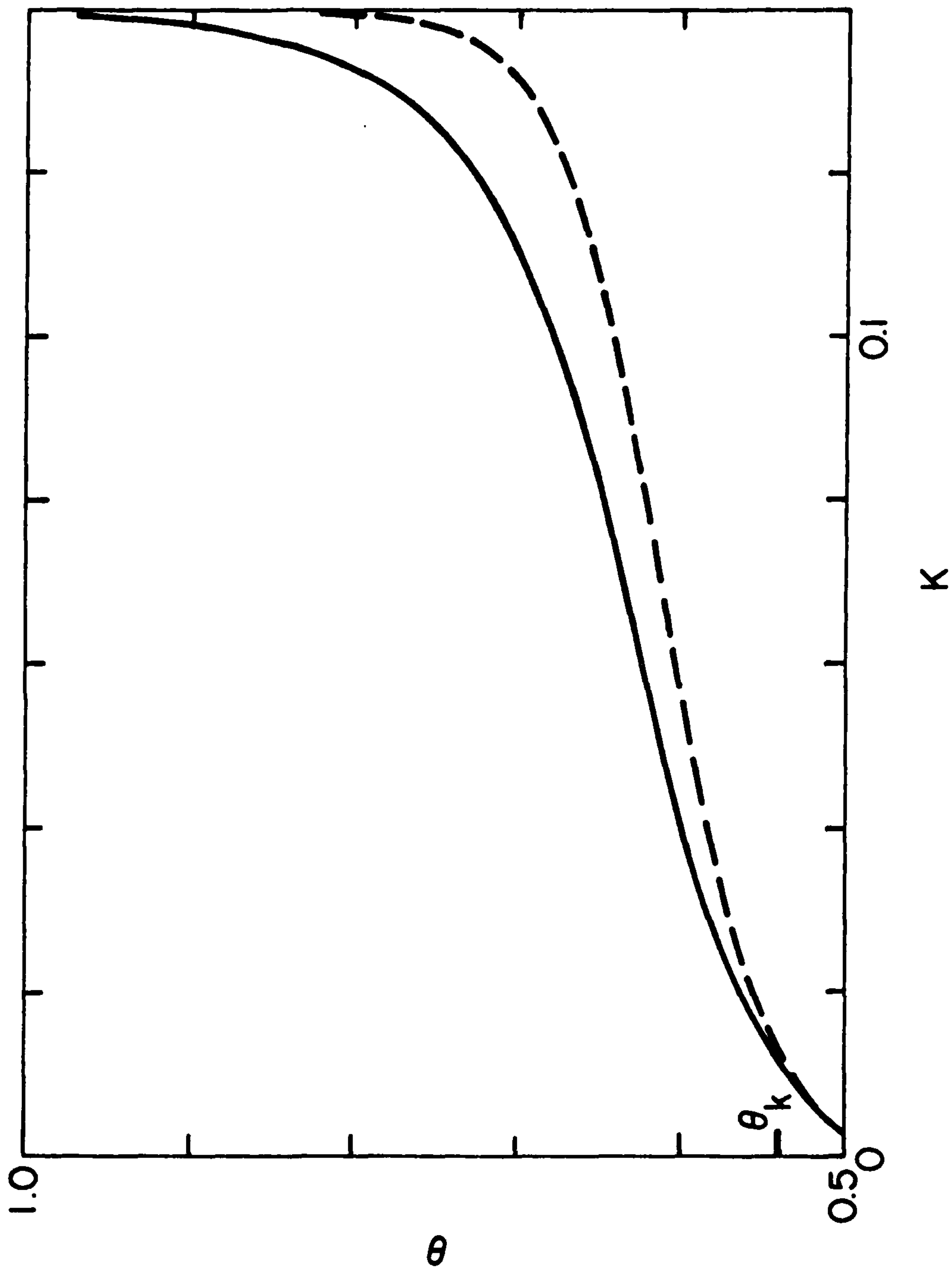

Figure 4. The transition from inert of diffusion-limited behavior for $\epsilon = 0.060$ (see Fig. 1 for legend).

derivative is infinite and defines one side of a cusp-like variation of $\theta(\tau)$. Values of practical interest imply that $m = 0(1) > 0$ and we therefore assume this inequality prevails. The implication is that as complete consumption of the particle is approached its temperature increases.

The numerical solution corresponding to $\epsilon = 0.060$ and shown in Fig. 1 indicates the nature of the $0(1)$ transition made by θ from θ_k to θ^*. The temperature parameter $\theta^* = 1.75$ in the present calculations. It is essential that $\theta^* > 1$ in the limit $\epsilon \to 0$ in order for cusp-like behavior to prevail. We note from the third of Eqs. 16 that the exponential term is exponentially small as $\epsilon \to 0$ provided $\theta^* > \theta_k$. It follows that R must likewise be small before the mass loss rate decreases from its diffusion-limited value. This behavior is typical of systems involving high activation energies where the reaction once initiated proceeds vigorously until the reactant supply (in the present case proportional to the surface area of the particle) is nearly eliminated. This argument is consistent with our earlier discussion of Eqs. 3 and 7.

On physical grounds we expect the extinction period to involve a reversal of the sign of the slope of $\theta(\tau)$ from the positive value prevailing at the end of the diffusion-limited period. During this period θ declines from θ^* to 1^+ while K decreases from K_d toward zero. We find that this period, although brief, involves a complex structure which is more conveniently analyzed by a reformulation of the basic equations as follows: If $R = R(K, \theta)$ is found from Eq. 3 and used to convert Eq. 2 into the equation

$$\frac{\partial R}{\partial K} K'(\tau) + \frac{\partial R}{\partial \theta} \theta' = -\frac{K}{R} \tag{21}$$

we obtain the single equation for $\theta = \theta(K)$

$$\frac{d\theta}{dK} = -\frac{\theta' R \dfrac{\partial R}{\partial K}}{K + R \dfrac{\partial R}{\partial \theta} \theta'}$$

which becomes after some calculation

$$\frac{d\theta}{dK} = -\frac{3\left[A\theta + D + \phi_r R(1-\theta^4)\right]\left[1 + K(1 - \exp(K - K_d))^{-1}\right]}{K^2 - (3K\theta_k/\epsilon\theta^2)(A\theta + D + \phi_r R(1-\theta^4))} \tag{22}$$

If an appropriate solution to Eq. 22 is obtained, Eq. 21 supplemented by $R = R(K, \theta\,(K))$, yields $K(\tau)$ and hence in terms of K as a parameter the other time histories, $R(\tau)$ and $\theta(\tau)$.

We show in Figs. 5 and 6 the exact numerical solutions in terms of $\theta(K)$ and $R(K)$ for $\epsilon = 0.060$. As time increases K decreases from its limiting value $K_d = 0.140$. The radius parameter R decreases monotonically toward zero while the temperature parameter θ exhibits the more complex behavior suggested earlier. Our task is to provide an asymptotic analysis approximating these characteristics as $\epsilon \to 0$.

To construct a description of the first portion of the extinction period, denoted Zone 1, we follow a two-step procedure; first Eqs. 18 and 20 suggest the forms

$$\theta \simeq \theta^* \,(1 - \chi(\epsilon)\,\bar{\theta} + \cdots) \tag{23}$$

$$K \simeq K_d \,(1 - \chi(\epsilon)\,\bar{K} + \cdots)$$

It follows from Eq. 3 that if $\chi(\epsilon)\bar{\theta} << 0(\epsilon)$

$$R \simeq \chi(\epsilon)/K_d \bar{K} \tag{24}$$

The importance of the inequality on χ is emphasized. As a second step the form of $\chi(\epsilon)$ is found by retaining in Eq. 1 the second terms in the expansions of $A(K)$ and $D(K)$ about $K = K_d$ and by requiring that these terms, previously neglected, be retained as $\epsilon \to 0$. As a result we find a distinguished limit if

$$\chi = (K_d/\tilde{\mu}\,\epsilon)^{1/2} \exp\left\{-\frac{1}{2\epsilon}\left[1 - \frac{\theta_k}{\theta^*}\right]\right\} \tag{25}$$

We note that this result indicates that the perturbations in Eq. 23 and in the particle size given by Eq. 24 are exponentially small in the limit $\epsilon \to 0$ provided θ^* is suitably large. This restriction is respected for sufficiently small values of ϵ, e.g., for $\epsilon = 0.060$. The different behavior near extinction

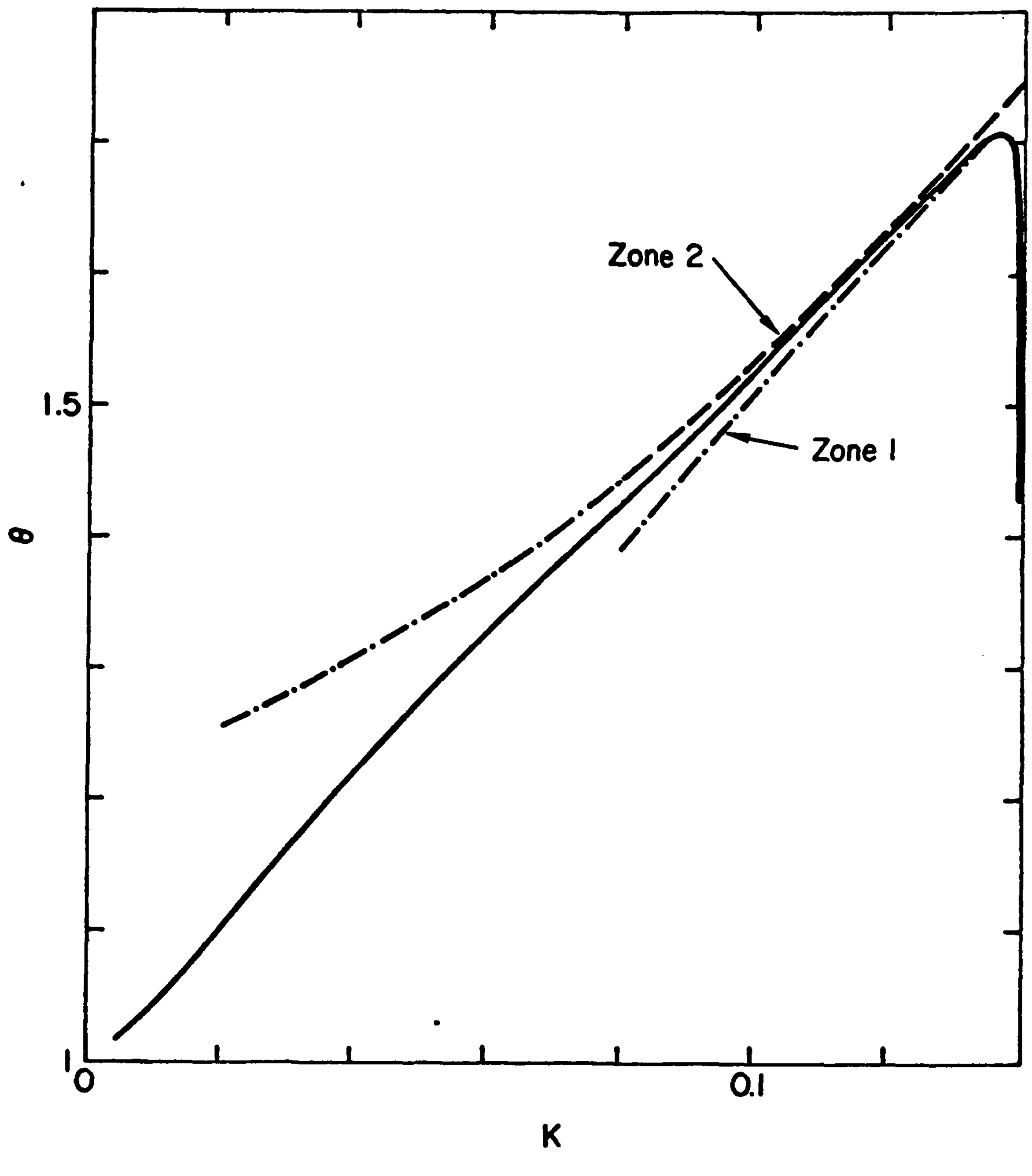

Figure 5. The temperature history during the extinction period: $\epsilon = 0.060$; ———— exact numerical solution; — — — asymptotic solution within the range of their validity; —·—·— asymptotic solution beyond the range of their validity.

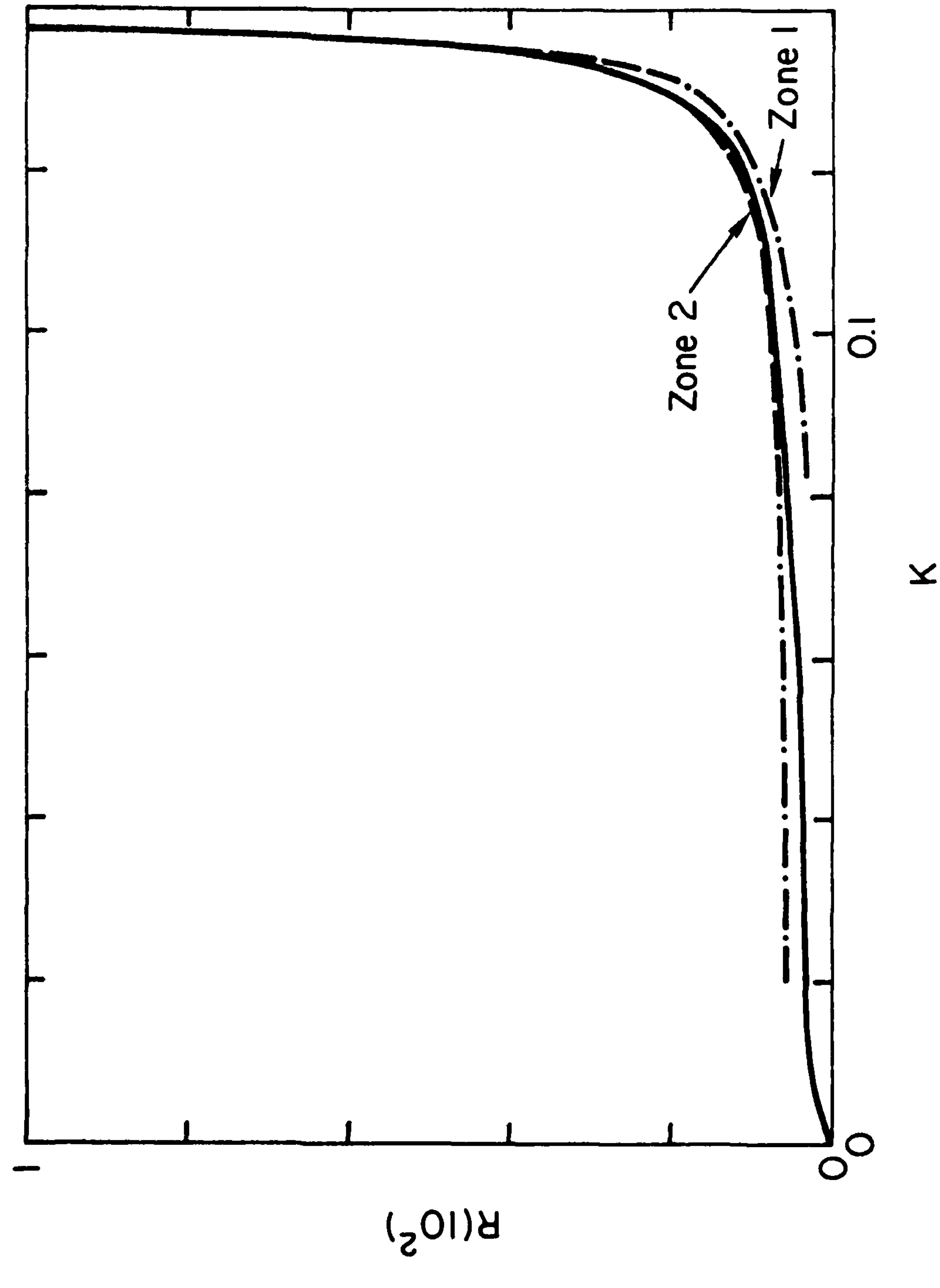

Figure 6. The radius history during the extinction period (see Fig. 5 for legend).

shown in Figs. 1-3 for the two values of ϵ suggests as might be expected that the asymptotic analysis is inapplicable for the larger value near extinction as well as earlier in the particle lifetime.

With Eqs. 23 and 25 substituted into Eq. 22 we obtain

$$\frac{d\bar{\theta}}{dK} + a\,\frac{\bar{\theta}}{\bar{K}} \simeq \frac{3}{\theta^*}\left[C_d + \frac{\phi_r(\theta^{*4}-1)}{K_d^3\bar{K}^2}\right] \tag{26}$$

where $C_d \equiv A'(K-K_d)\theta^* + D'(K-K_d) - \tilde{c}_p K_d e^{-K_d}(\theta^*-1)/(1-e^{-K_d})^2$ and $a \equiv 3\,A_d/K_d$ are positive constants. To match the solution of Eq. 26 to the behavior of Eq. 20 as $s \approx 1/2^-$ requires

$$\bar{\theta}(\bar{K} \to 0) \approx m/(\theta^* K_d \bar{K})$$

The solution of Eq. 26 is

$$\bar{\theta} = \frac{3\,C_d}{\theta^*(a+1)}\,\bar{K} + \frac{3\phi_r(\theta^{*4}-1)}{\theta^*(a-1)\,K_d^3\bar{K}} \tag{27}$$

In Eq. 27 a complementary solution K^{-a} is suppressed since with $a > 1$, the required behavior as $\bar{K} \to 0$ prohibits its retention. The coefficient of $\bar{K}^{-1}$ equals $(m/\theta^* K_d)$ so that matching is assured.

The θ-solution obtained from Eqs. 23 and 27 describes the approach with increases in time toward θ^* and its departure therefrom as $\bar{K}$ increases. The concomitant decline in the radius is obtained from Eq. 24. Consequently the solution which rounds off the cusp-like behavior given by the preceding period takes place on a small time scale defined by $\tau - (\tau_k + (1/2K_d) + \lambda^{-2}\epsilon \ln(1/\epsilon)) = 0(\chi^2(\epsilon))$.

We show on Figs. 5 and 6 the asymptotic solutions as dashed lines within the range of validity of K and extend them beyond that range as dot-dashed lines. When this procedure is followed for the solutions given by Eqs. 27 and 24, we find that within plotting accuracy the asymptotic and numerical solutions cannot be distinguished within the range of validity of the former. As a consequence only the extensions of these solutions for Zone 1 appear on Figs. 5 and 6.

To find the solution in the layer denoted Zone 2 let

$$\theta \simeq \theta^{*}\,(1 - \epsilon\, g(k) + \cdots) \tag{28}$$

$$K \simeq K_d\,(1 - \epsilon\, k + \cdots)$$

As a consequence of these forms

$$R \simeq \chi^2 \exp\,(\theta_k\, g/\theta^{*})/\epsilon\, K_d\, k$$

which indicates a further dramatic reduction in particle radius. Then Eq. 22 becomes

$$\frac{dg}{dk} \simeq - \frac{ag - (3\,C_d/\theta^{*})\,k}{k\left[1 - (\theta_k/\theta^{*})\,(ag - (3\,C_d/\theta^{*})\,k\right]} \tag{29}$$

which is to be solved subject to the matching condition $g(k \to 0) \to 0$.

For increasing values of k corresponding to increasing time the asymptotic form of Eq. 29 is

$$\frac{dg}{dk} \simeq \frac{\theta^{*}}{\theta_k\, k}$$

which has the solution

$$g(k \to \infty) \simeq \frac{\theta^{*}}{\theta_k} \ln k + g_o \quad . \tag{30}$$

It follows from Eqs. 28 and 30 that the solution in this zone is singular when $\epsilon k = 0\ (1)$ which corresponds to $\epsilon g = 0\ (\epsilon \ln (1/\epsilon))$ and $R = 0\ (\chi^2/\epsilon)$. Significantly, the nonuniformity with respect to K suggests that the mass loss declines significantly from K_d in the next zone.

Numerical solutions to Eq. 29 and to the related solution for $R = R(K)$ are added to Figs. 5 and 6 to permit comparison with the exact numerical results. Again the asymptotic solutions are continued by dot-dash lines beyond an appropriate value of K to indicate their range of validity. Satisfactory agreement between the two solutions is achieved.

We find that in the next zone, Zone 3, in which K declines rapidly we must take

$$\theta \simeq \theta^* \left[1 - \frac{\theta^*}{\theta_k} \epsilon \ln\left(\frac{1}{\epsilon}\right) - \epsilon h(K) + \cdots \right] \tag{31}$$

The equation for $h(K)$ is

$$\frac{dh}{dK} \simeq -\frac{\theta^*}{\theta_k} \left[\frac{1}{K} + \frac{1}{1 - \exp(K - K_d)}\right] \tag{32}$$

which is to be solved subject to the matching condition

$$h(K \to K_d^-) \simeq -\frac{\theta^*}{\theta_k} \ln\left[\frac{K_d}{K_d - K}\right] + g_o$$

The 0(1) constant of integration g_o is obtained from the K-solution (Eq. 29). The solution of Eq. 32 is

$$h = g_o - \frac{\theta^*}{\theta_k} \left\{\ln\left[K - \ln \exp\left[(K_d - K) - 1\right]\right]\right\} \tag{33}$$

When comparison between the asymptotic and numerical solutions is considered, we encounter a practical limitation of the former solutions. For the values of the parameters used for the numerical solutions we find that the second term on the right side of Eq. 31 is 0.54, indicating that the value of $\epsilon = 0.060$ overstrains the asymptotic analysis. Accordingly, we cease making comparison of the two solutions but for completeness carry through the asymptotic analysis with the expectation that agreement with numerical solutions would be achieved for suitably small values of ϵ. For example, with all other parameters fixed at their present values a value of $\epsilon = 0.022$ would make the second term in Eq. 31 equal to 0.1, an appropriately small value.

When Eq. 33 is substituted into Eq. 32, we find that a nonuniformity occurs when $(\theta^*/\theta_k)\epsilon \ln(1/K) = 0(1)$ implying that the mass loss rate becomes vanishingly small at the end of this zone. As a consequence the temperature decreases further from θ^*. Physically, the chemical reaction weakens, permitting convective cooling to reduce the temperature of the dying particle so that $\theta \to 1$. Also in this zone Eq. 3 indicates that R remains nearly constant at a small magnitude of $0(\chi^2/\epsilon)$.

The major decline in θ toward unity occurs in Zone 4 in which it is found convenient to scale K by the transformation

$$K = \frac{1}{\epsilon} \exp\left[-\frac{\theta_k p}{\theta^* \epsilon}\right] \tag{34}$$

which defines p as a new $0(1)$ independent variable. This exponential transformation is suggested by the nonuniform behavior of the solution in the preceding zone. Then to all algebraic orders in ϵ the θ-equation is simply

$$\frac{d\theta}{dp} = -\frac{\theta^2}{\theta^*} \tag{35}$$

$$\theta(p \to 0) \simeq \theta^*$$

where the latter is the appropriate matching condition. The solution of Eq. 35 is

$$\theta = \frac{\theta^*}{1+p} \qquad 0 < p < (\theta^* - 1)$$

where the limit on p is required to prevent the temperature from dropping below unity. During this cooling phase the exponentially small radius is basically constant and given by

$$R \simeq R_o = \frac{1}{\epsilon^2 Y_{1\infty}} \exp\left[-\frac{1}{\epsilon}\left[1 - \frac{\theta_k}{\theta^*}\right]\right] \tag{36}$$

These results can be used in an appropriate form of Eq. 1 to show the time-dependence of the temperature decline; we find that

$$\theta = 1 + (\theta^* - 1) \exp\left[-\frac{3\tilde{C}_p}{R_o^2}(\tau - \tau^*)\right] \tag{37}$$

where $\tau^* = \tau_k + (1/2K_d) + \lambda^{-2}\epsilon \ln (1/\epsilon)$. The small variation from the extinction time should be noted, the extremely brief time scale of the major temperature decline being implicitly indicated by the exponent in Eq. 37. This result is valid for all algebraically small orders of ϵ.

Further decay of a minute nature occurs as $R \to 0$ and $\theta \to 1$. This zone provides little additional information on particle behavior and is not considered further.

CONCLUDING REMARKS

The method of activation energy asymptotics is applied to the burning of carbon particles in a hot oxiding ambient. To simplify the analysis gas phase reactions are assumed negligible and only the direct heterogeneous reaction between carbon and oxygen is taken into account. It is shown that a complete description of particle history is largely given by an induction period during which the particle is heated to an ignition temperature and by a subsequent period during which the rate of mass loss is diffusion-limited. Such a picture is not dissimilar to earlier approximate calculations of particle behavior based on ad hoc assumptions. However, the present analysis provides clear indications of the conditions under which the analysis applies. In fact a significant portion of our discussion relates to the applicability to a specific calculation involving ϵ small compared to unity but nonzero of the limiting behavior arising when the small parameter $\epsilon \to 0$. The asymptotic analysis provides estimates for quantities of applied interest: conditions under which ignition occurs, conditions for diffusion-limited mass loss and the times for ignition and complete consumption.

The results of two representative numerical examples, one respecting the limits of applicability of the asymptotic analysis and a second overextending that analysis, are compared with the predictions of AEA. Good agreement is found for the former while as expected significant disagreement is found for the latter.

During a brief extinction period all of the quantities describing particle behavior undergo significant variations. As a consequence the asymptotic solutions for this period are complex and impose significant limitations on the acceptable size of the expansion parameter. In fact only two of the four zones found to be required are compared to an exact numerical solutions.

Extension of the present analysis to include gas phase reactions and more complex composition in the surrounding ambient can be carried out at the expense of increased algebraic complexity. However, the main features of the analysis of applied interest, namely the description of the induction and a diffusion-limited periods, may involve straight-forward considerations.

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NOMENCLATURE

a, C_d, m	parameters arising in the extinction period
$A(K), D(K)$	functions of mass loss parameter
$\tilde{c}_p$	nondimensional parameter related to the coefficient of specific heat at constant pressure
E	thermochemical parameter
g, h	perturbation functions arising in extinction period
K	mass loss parameter
K_d	diffusion-limited value of K
$\tilde{K}_1$	nondimensional reaction rate parameter
k	perturbed mass-loss parameter
r_p	instantaneous particle radius
r_{po}	initial particle radius
R	nondimensional radius parameter
s	transformed time variable
T	temperature
T_a	activation temperature
T_∞	temperature in surrounding ambient
$Y_{1\infty}$	oxygen concentration in surrounding ambient

Greek Symbols

α	temperature ratio, T_∞/T_a
$\tilde{\Delta}, \tilde{\Delta}_s$	thermochemical parameters

θ	nondimensional temperature parameter
$\theta_k, \theta_k^*, \theta^*$	nondimensional temperature parameters arising in AEA
ϵ	expansion parameter
$\tilde{\mu}, \hat{\mu}, \mu'$	molecular weight parameters
ϕ_r	radiative parameter
λ^2, ω^2	parameters in AEA
τ	nondimensional time
$\tilde{\tau}$	transformed time variable

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The method of activation energy asymptotics is used to describe the entire history of a carbon particle suddenly immersed in a hot oxidizing ambient. Under appropriate conditions which are established by the analysis the history of the particle is shown to consist principally of a heat-up period during which no significant chemical reaction takes place followed by a period involving diffusion-limited mass loss and terminating in complete

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ABSTRACT (continued)

particle consumption. Between these two extended periods brief ignition and post-ignition periods are described. The final demise of the particle occurs in a complex extinction period during which all the quantities describing the particle behavior undergo large variations.

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